

What we claim is:

5 1. A modified alkali silicate matrix composition comprising the reaction product of an alkali silicate and/or alkali silicate precursors, one or more acidic oxoanionic compounds, water and optionally one or more compounds containing multivalent cation(s) selected from Groups 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15 or 16 of the periodic table and optionally a clay and/or oxide filler.

10 2. The composition of claim 1, wherein the alkali silicate is selected from potassium silicate solutions and/or sodium silicate solutions.

15 3. The composition of claim 1, wherein the alkali silicate is crystalline and/or amorphous sodium silicate and/or potassium silicate.

4. The composition of claim 1 wherein the alkali silicate precursors are an alkali base and a SiO_2 source.

20 5. The composition of claim 4 wherein the SiO_2 source is an amorphous or crystalline SiO_2 form, selected from the group consisting of silica, silica fume, microsilica, precipitated silica, sand, quartz, quartz flour, silica gels, fumed silica and colloidal silica.

25 6. The composition of claim 4 wherein the alkali base is an alkali hydroxide, alkali carbonate and/or an alkali oxide.

7. The composition of claim 6 wherein the alkali hydroxide is selected from the group consisting of potassium hydroxide and sodium hydroxide.

8. The composition of claim 1 wherein the alkali silicate and/or alkali silicate precursors has a $\text{SiO}_2/\text{A}_2\text{O}$ ratio of about 2.0:1.0 to 20.0:1.0, where A is K (potassium) and/or Na (sodium).

5 9. The composition of claim 1, wherein said acidic oxoanionic compound is selected from the group of consisting of boric acid, phosphoric acid, sulfuric acid, nitric acid, sodium dihydrogen phosphate, disodium hydrogen phosphate, their potassium analogs, their ammonium analogs, and mixtures thereof.

10 10. The composition of claim 1, wherein the acidic oxoanionic compound is phosphoric acid.

15 11. The composition of claim 1, wherein the compounds containing multivalent cation(s) are selected from the group compounds incorporating boron, aluminum, lead, zinc, gallium, cadmium, titanium, zirconium, lanthanum, cerium, neodymium, yttrium, calcium, magnesium, strontium and barium.

20 12. The composition of claim 1, wherein the compounds containing multivalent cation(s) are selected from the group consisting of magnesium nitrate, magnesium sulfate, magnesium chloride, zinc nitrate, zinc sulfate and zinc chloride.

 13. The composition of claim 1 wherein the compounds containing multivalent cation(s) is zinc chloride.

25 14. The composition of claim 1 wherein the clay filler is kaolin, calcined kaolin, mica and/or metakaolin.

30 15. The composition of claim 1, wherein said alkali silicate and/or alkali silicate precursors are present in an amount between about 30 wt. % and 90 wt. % based upon the total composition.

16. The composition of claim 1, wherein said alkali silicate is formed from a alkali hydroxide and a SiO_2 source and said alkali hydroxide is present in an amount of between about 3 wt. % to about 25 wt. % and said SiO_2 source is present in an amount of about 15 wt. % to about 75 wt. % based upon the total composition weight.

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17. The composition of claim 1, wherein said acidic oxoanionic compound is present in an amount of between about 0.01 wt. % and about 20 wt. % based upon the total composition.

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18. The composition of claim 1, wherein said compounds containing multivalent cation(s) is present in an amount of between zero and about 20 wt. % based upon the total composition.

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19. The composition of claim 1, wherein said clay filler is present in an amount of between about 0.0 wt. % and about 25 wt. % based upon the total composition weight.

20. The composition of claim 1 wherein the network comprises the reaction product of the following:

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- a) 30 to 90 wt. % of alkali silicate,
- b) 0.01 to 20 wt. % of phosphoric acid,
- c) 0 to 25 wt. % of a clay filler,
- d) 0 to 20 wt. % of at least one multivalent metal salt and
- e) 10 to 70 wt. % of water.

21. The composition of claim 1, wherein the network comprises the reaction product of the following:

- a) 10 to 75 wt. % of a SiO_2 source,
- 5 b) 3 to 25 wt. % of an alkali hydroxide,
- c) 0.01 to 20 wt. % of phosphoric acid,
- d) 0 to 25 wt. % of a clay filler,
- e) 0 to 20 wt. % of at least one multivalent metal salt and
- f) 10 to 70 wt. % of water.

22. The composition of claim 1 wherein the network comprises the reaction product of the following (excluding water):

- a) 30 to 90 wt. % of alkali silicate,
- b) 0.01 to 30 wt. % of phosphoric acid,
- 15 c) 0 to 30 wt. % of a clay filler and
- d) 0 to 25 wt. % of at least one multivalent metal salt.

23. The composition of claim 1 wherein the network comprises the reaction product of the following (excluding water):

- 20 a) 10 to 90 wt. % of SiO_2 source,
- b) 3 to 25 wt. % of alkali hydroxide,
- c) 0.01 to 30 wt. % of phosphoric acid,
- d) 0 to 30 wt. % of a clay filler and
- e) 0 to 25 wt. % of at least one multivalent metal salt.

24. The composition of claim 1, wherein the network also includes non-clay filler materials.

25. The composition of claim 24, wherein the filler materials are selected from the group consisting of fibers, spheres, and particles.

26. The composition of claim 25, wherein the fibers are selected from a group consisting of continuous fibers and discontinuous fibers.

27. The composition of claim 25, wherein the spheres are selected from the group consisting of microspheres, macrospheres, hollow spheres, and solid spheres comprising glass, ceramic, metal, mineral, organic or inorganic materials.

28. The composition of claim 25, wherein the filler materials are utilized for reinforcement and selected from the group consisting of carbon fibers, glass fibers, alkali resistant glass fibers, organic fibers, ceramic fibers, mineral fibers and metallic fibers.

29. The composition of claim 28, wherein the reinforcement materials are selected from the group consisting of graphite fibers, E-glass fibers, S-glass fibers, basalt fibers, stainless steel fibers, titanium fibers, nickel alloy fibers, aramid fibers, polyethylene fibers, SiC fibers and BN fibers.

30. The inorganic matrix composition of claim 1 formed by curing the composition at a temperature ranging from about 15°C to 1000°C+, an external pressure from ambient to about 20,000 psi, and optionally under a vacuum of from about ambient to about 10^{-3} torr.

31. The inorganic matrix composition of claim 1 formed by curing the composition at about 15°C to about 200°C and an external pressure from ambient to 200 psi, and optionally under a vacuum of from about ambient to 10^{-3} torr.

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32. A method of making an inorganic silicate composite comprising the steps of applying an aqueous slurry of a mixture of an alkali silicate, phosphoric acid, a clay filler, an alkaline earth salt and water to a reinforcing medium and curing the composite by applying heat at a temperature of about 15°C up to about 1000°C+, an external pressure from ambient to about 20,000 psi and under, and optionally under a vacuum of about ambient to about 10^{-3} torr.

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33. The method of claim 32 wherein the composite is cured preferably at a temperature of about 15°C up to about 200°C and an external pressure from ambient to 200 psi, and optionally under a vacuum of about ambient to about 10^{-3} torr.

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34. The method of claim 32 wherein the uncured composite is in form of bulk molding compound, sheet molding compound, powder and reinforcement, liquid and reinforcement or prepreg.

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35. The method of claim 32 wherein the composite is cured using processes which include compression molding, pultrusion, wet lay-up, filament winding, autoclave vacuum bag processing, non-autoclave vacuum bag processing, vacuum infusion, liquid resin, film infusion or powder infusion, resin transfer molding, extrusion, injection molding, casting, spin casting, trapped elastomer molding and like processes.

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36. The method of claim 32 wherein the composite is consolidated before and/or during the curing process.

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37. The method of claim 32 wherein the slurry includes additional filler materials selected from the group consisting of carbon fibers, glass fibers, alkali resistant fibers, mineral fibers, organic fibers, ceramic fibers and metallic fibers.

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38. The method of claim 32 wherein the slurry includes additional filler materials selected from the group consisting of graphite fibers, E-glass fibers, S-glass fibers, stainless steel fibers, titanium fibers, nickel alloy fibers, aramid fibers, polyethylene fibers, basalt fibers, SiC fibers and BN fibers.

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39. The method of claim 32 wherein the reinforcing medium is a glass fiber such as an E-glass fiber, S-glass fiber and/or an alkaline resistant fiber.

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40. The method of claim 32 wherein the slurry further includes a surface-active agent.

41. The method of claim 32 wherein the slurry further includes an anionic,
5 cationic and/or a nonionic surface-active agent.

42. The method of claim 32 wherein the slurry further includes a surface-active agent selected from the group consisting of alkylaryl sulfonates, silicones, quaternary ammonium salts, protonated organoamine salts, and combinations thereof.

10 43. The method of claim 32 wherein the slurry further includes a surface-active agent in an amount from none to 10 wt. % based upon the weight of the total composition.

15 44. The method of claim 32 wherein the slurry further includes a surface-active agent in an amount from 0.5 wt. % to 5 wt. %. based upon the weight of the total composition.

20 45. An alkali silicate slurry composition for forming an inorganic polymer network comprising an alkali silicate or a mixture that yields an insitu alkali silicate, an acidic oxoanionic compound, water and optionally one or more compounds containing multivalent cation(s) selected from Groups 2, 3, 4, 5, 6, 7, 8, 9,10, 11, 12, 13, 14, 15 or 16 of the periodic table and optionally a modifier that moderates the gelation of matrix binder suspension and/or optionally a clay filler to enhance physical and thermal properties.

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46. The composition of claim 45, wherein the gel modifier is an organic acid or organic base.

30 47. The composition of claim 45, wherein said organic acid is an α -hydroxyacid and/or a β -hydroxyacid.

48. The composition of claim 45, wherein said organic base is substituted nitrogen organic base.

49. The composition of claim 45, wherein said organic base is a substituted
5 pyridine.

50. The composition of claim 45, wherein said organic base is quinoline.

51. The composition of claim 45, wherein said gel modifier is present in an
10 amount of between zero and about 10 wt. % based upon the total composition.

52. A method of making an inorganic silicate composite comprising the steps
of applying an aqueous slurry of a mixture of an alkali silicate, phosphoric acid, water
and optionally a clay filler, an alkaline earth salt and/or a gel modifier to a reinforcing
15 medium and curing the composite by applying heat at a temperature of about 15°C up to
about 1000°C and external pressures from ambient pressure to about 20,000 psi and/or
under a vacuum of from about ambient to about 10^{-3} torr.